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(71)Applicant :

NISSAN MOTOR CO LTD

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(72)Inventor:

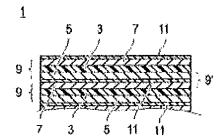
OSAWA YASUHIKO

HISAMITSU YASUNARI

### (54) BIPOLAR BATTERY

## (57)Abstract:

PROBLEM TO BE SOLVED: To provide a bipolar battery made by using a novel collector excellent in corrosion resistance at a positive electrode side and not alloying with lithium at a negative electrode side, with its thickness appropriate in comparison with an electrode layer and an electrolyte layer. SOLUTION: Of the bipolar battery 1 having a structure of laminating electrode lamination bodies formed by laminating a positive electrode layer 5 on one face of a sheet of polymer electrolyte film 3 and a negative electrode layer 7 on the other face through an electron conductive layer, the electron conductive layer 11 contains mainly an insulating polymer and an electron conductive filler.



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#### CLAIMS

[Claim(s)]

[Claim 1]

In a bipolar battery with structure which laminated an electrode layered product which has a positive electrode layer on a field of one of the two of polymer electrolyte membrane of one sheet, and has a negative electrode layer on a field of another side via a laver of electron conductivity.

A bipolar battery, wherein a electron conductive layer mainly contains an insulating polymer and a electron conductive filler.

[Claim 2]

In a bipolar battery with structure which laminated an electrode layered product which has a positive electrode layer on a field of one of the two of polymer electrolyte membrane of one sheet, and has a negative electrode layer on a field of another side via a layer of electron conductivity.

A bipolar battery with which a electron conductive layer is characterized by having a layer which mainly contains an insulating polymer and a electron conductive filler at least in one side of a metal layer.

[Claim 3]

The bipolar battery according to claim 1 or 2, wherein a electron conductive filler is carbon black.

[Claim 4]

A cell group which connects two or more bipolar batteries of a description to any 1 clause of Claims 1-3, and is characterized by things.

[Claim 5]

Vehicles which carry a bipolar battery and/or the cell group according to claim 4 of a description in any 1 clause of Claims 1-3, and are characterized by things.

[Translation done.]

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## **DETAILED DESCRIPTION**

[Detailed Description of the Invention]

[0001]

[Field of the Invention]

This invention relates to the bipolar battery with which it comes to arrange a positive electrode layer and a negative electrode layer at the both sides of a charge collector. [0002]

[Description of the Prior Art]

In recent years, the rechargeable lithium-ion battery which can attain high energy density and high power density has been developed as large capacity power supplies, such as an electromobile. The cathode in which the basic constitution of the rechargeable lithium-ion battery used the binder for the aluminum collector, and applied conductive auxiliaries, such as positive active material, such as cobalt acid lithium, and acetylene black, to it, The anode which used the binder for the copper charge collector and applied carbon particulates to it is arranged via the porous membrane separator of a polyolefin system, and it has become what filled the nonaqueous electrolyte which contains LiPF<sub>6</sub> etc. in this. In applying to an electromobile etc., the series connection of the cell (cell) of this composition is carried out, and the series connection of a battery-module unit and also the module is carried out, and it constitutes and uses a cell group.

From a viewpoint of the energy density of a cell, and power density, that connection between cells and inter module connection are [ connection resistance ] spatial and weight improvement are desired. Recently, the proposal of the cell which adopted the bipolar electrode unit which resistance reduction of connection between this cell is possible, and can expect miniaturization was made (for example, refer to patent documents 1.). in this proposal, strip processing of two kinds of metallic foils was carried out to the charge collector — since the gel electrolyte is used for the electrolyte using the clad plate so to speak, the sealing seal in each cell unit is indispensable, and the liquid junction between cells may start. If a solid polymer electrolyte is used instead of this gel electrolyte, a sealing seal becomes unnecessary and a realistic bipolar battery can be constituted (for example, the patent documents 2, three references.). In the bipolar battery which are all the solid polymer cells constituted using a polymer electrolyte as this electrolyte, since an electrolysis solution is not included, there is neither liquid leakage nor the generation of gas, and it has the advantage that it is very reliable. (For example, the patent documents 2, three references.).

[0004]

[Patent documents 1]

JP,H11-204136,A

[Patent documents 2]

JP,2000-100471,A

[Patent documents 3]

JP,2002-75455,A

[0005]

[Problem to be solved by the invention]

However, since the conventional bipolar battery used a metallic foil and its layered product for the collector layer, the charge collector which can be used was restricted very much for alloying with lithium in the corrosion-resistant problem and anode by the side of a cathode. A negative electrode layer is allotted to one side of current collecting foil about tens of micrometers thick in the field of a positive electrode layer and another side, it is the structure which laminated this via the high polymer electrolyte layer, the thickness of current collecting foil becomes thick compared with an electrode layer and an electrolyte layer, and the energy density of a cell and power density decline.

[0006]

Therefore, it is a new charge collector which this invention excels [ place / target ] in the corrosion resistance by the side of a cathode, and does not produce alloying with lithium in an anode, and is providing the bipolar battery which uses a charge collector with the proper thickness compared with an electrode layer and an electrolyte layer.
[0007]

[Means for solving problem]

In a bipolar battery with the structure which laminated the electrode layered product which this invention has a positive electrode layer on the field of one of the two of polymer electrolyte membrane of one sheet, and has a negative electrode layer on the field of another side via the layer of electron conductivity, it is a bipolar battery, wherein a electron conductive layer mainly contains an insulating polymer and a electron conductive filler.

[8000]

[Effect of the Invention]

In the bipolar battery of this invention, the things (an insulating polymer, carbon black, etc.) which are not metallic foils can be used for the electron conductive layer as a charge collector, Excel in the corrosion resistance by the side of a cathode, and alloying with lithium in an anode is not produced, Furthermore a electron conductive layer can be made thinner than the case where a tens of micrometers (usually about 35 micrometers) metallic foil is used by the usual method, this electron conductive layer thickness can be made into the proper range (about 1-20 micrometers) to an electrode layer and an electrolyte layer (film), and the energy density of a cell and power density can be enlarged. Therefore, in various industries, it becomes a useful power source.

[0009]

[Mode for carrying out the invention]

Hereafter, it explains per embodiment of the invention.

[0010]

In a bipolar battery with the structure which laminated the electrode layered product which a first embodiment of the bipolar battery concerning this invention has a positive electrode layer on the field of one of the two of polymer electrolyte membrane of one sheet, and has a negative electrode layer on the field of another side via the layer of electron conductivity, A electron conductive layer mainly contains an insulating polymer and a electron conductive filler.

[0011]

Although a electron conductive layer is usually called charge collector (a bipolar battery cell interval wall and electronic conductor) and conductivity is using the high metallic foil electronically, since current flows into an electrode surface and a perpendicular direction in a bipolar battery, so high electron conductivity is necessarily unnecessary. Conventionally, as a manufacturing method of an electrode, electrode stock slurry was applied to the metallic foil, and dry solidification (it polymerizes depending on the case) was carried out to it. Therefore, a certain amount of intensity was required for it, and although the metallic foil to be used was based also on construction material from these things, a thickness of about 15-20 micrometers was inevitably required for it. So, in the first form of this invention, it is considered as the structure using the thin film which made the insulating polymer newly fill up with a electron conductive filler as this electron conductive layer taking advantage of the feature which is a bipolar battery.

[0012]

In a first embodiment of the bipolar battery concerning this invention. The electron conductivity whose thin film which made the insulating polymer fill up with a electron conductive filler as a electron conductive layer is so expensive is advantageous at the point which can choose the raw material which slurrying of a raw material is easy and can carry out [ thin film ]-izing to about several micrometers simply from knowledge that it is not necessarily required. Namely, the electron conductive layer can use now construction material other than metal for the electron conductive layer as a charge collector, and using especially carbon as a electron conductive filler, By applying the stock slurry for electron conductive layers other than attachment of this electron conductive layer (thin layer) directly, a electron conductive layer can be formed and a bipolar battery with high energy density and power density can be constituted. In the point which can choose material without the corrosion-resistant problem by the side of a cathode, or the problem of alloying with lithium in an anode as compared with a metallic foil, it is advantageous. Since slurrying of a raw material can choose an easy material, apply electrode stock slurry to both sides of the charge collector which has also structurally fixed intensity, and an electrode is not formed in them, For example, it is advantageous at the point which can apply the stock slurry of a electron conductive layer, can carry out dry solidification, and can form a electron conductive layer on the electrode which applied electrode stock slurry to both sides of a high polymer electrolyte layer with fixed intensity, formed the electrode, and was formed in both sides of this high polymer electrolyte layer. That is, in order for what is necessary to be just to form a electron conductive layer on an electrode contrary to the former, this electron conductive layer is very advantageous also in the point which does not need at all intensity which was needed by the conventional manufacturing process, but can carry out [ thin film ]-izing simply.

As for electron conductive layer thickness, in a first embodiment of the bipolar battery concerning this invention, it is desirable for 1-20 micrometers to be preferred and to consider it as the range of 5-10 micrometers more preferably from a viewpoint [ thin-film-ize and ] of enlarging the energy density of a cell and power density. When there is possibility of an ion short circuit when electron conductive layer thickness is less than 1 micrometer, and exceeding 20 micrometers, a merit is small in energy density.

[0014]

Where a electron conductive filler is distributed in an insulating polymer, ion conduction resistance should just set [ very ] and kick greatly [ electronic conduction resistance ] the electron conductive layer which can be used by a first embodiment of the bipolar battery concerning this invention to a plane direction.

Among these, especially as the above-mentioned electron conductive filler, it should not be restricted, and it can be used conventionally, choosing from a publicly known thing suitably, and carbon black, metal particles, electrically conductive ceramics, etc. can be used. Especially, from the dispersibility into the insulating polymer of the above-mentioned electron conductive layer, it is a point which can perform various surface modifications, and it can be said that carbon black is preferred. By using carbon black for the electron conductive filler of a electron conductive layer, the purpose (high-energydensity-izing, high-power-density-izing) can be more easily attained by low cost. [0016]

As for the size of the above-mentioned electron conductive filler, it is good that it is less than 0.5 micrometer from

viewpoints of dispersibility etc. In exceeding 0.5 micrometer, it becomes difficult to build a thin film. Let a size here be the average value of the particle diameter (size) of the particles (foundations) of a electron conductive filler. [0017]

Especially as an insulating polymer, it is not restricted, and it can be used, choosing from what [ as shown below ] is conventionally publicly known suitably for example.

[0018]

(1) A thermoplastic insulating polymer can be used. As this thermoplastic insulating polymer, polyethylene, polypropylene, polystyrene, etc. can use it conveniently, for example. [0019]

Although the method of forming the electron conductive layer which carried out kneading dispersion to the insulating polymer which carried out melting of the electron conductive filler, for example as a method of manufacturing a electron conductive layer using the insulating polymer of this thermoplasticity and which was made to thin-film-ize by extrusion-molding processing, etc. are mentioned, it should not be restricted to these at all. [0020]

(2) What the above-mentioned insulating polymer and a electron conductive filler can distribute to both solvents is usable. The insulating polymer of this solvent dispersibility does not affect battery capacity, and just distributes it to a solvent. As an insulating polymer of this solvent dispersibility, a neoprene/toluene system, and various fluoroelastomers / methyl-ethyl-ketone systems can be used, for example.

[0021]

As a method of manufacturing a electron conductive layer using the insulating polymer of this solvent dispersibility, For example, the stock slurry for electron conductive layers which distributes this insulating polymer and a electron conductive filler to a solvent is prepared, The thin film of a electron conductive layer can be produced by applying with direct spray coating and a doctor blade on the electrode which formed this stock slurry for electron conductive layers in both sides of the electrolyte layer which has fixed intensity, for example, respectively etc.

[0022]

After applying this stock slurry for electron conductive layers to a glass plate or a PET film, drying a solvent and producing a thin film, this thin film can be stuck on an electrode and the thin film of a electron conductive layer can also be formed by removing a glass plate and a PET film after that.
[0023]

Also in which process method of of the above (1) and (2), the content of electron conductive fillers, such as carbon black in the electron conductive layer obtained, here, it is preferably desirable five to 50 mass % and to adjust the loadings of the electron conductive filler in a raw material so that it may become the range of 10 - 30 mass % more preferably. When resistance becomes large at the case of less than 5 mass % and the content of a electron conductive filler exceeds 50 mass %, it becomes difficult to build a uniform thin film.

[0024] Hereafter a first t

Hereafter, a first typical embodiment of the bipolar battery concerning this invention is briefly described using Drawings. However, it cannot be overemphasized that it is not what should be restricted to the embodiment which requires this invention in any way.

[0025]

Drawing 1 shows the partial section schematic view which expressed typically a first embodiment of the bipolar battery concerning this invention. As shown in drawing 1, the bipolar battery 1 of this invention, It has the positive electrode layer 5 on the field of one of the two of polymer electrolyte membrane (high polymer electrolyte layer 3) of one sheet, It has the structure which carried out the plural laminates of the electrode layered product 9 which has the negative electrode layer 7 via the electron conductive layer 11 on the field of another side, and the electron conductive layer 11 mainly contains an insulating polymer and a electron conductive filler. In other words, the positive electrode layer 5 is arranged to the field of 1 of the electron conductive layer 11 as a charge collector, and it can be said that it has the structure which carried out the plural laminates of bipolar electrode 9' of the structure which has arranged the negative electrode layer 7 via polymer electrolyte membrane (high polymer electrolyte layer 3) in other fields. That is, the positive electrode layer 5, the electron conductive layer (charge collector) 11, and the negative electrode layer 7 have the structure laminated in this order. [0026]

What is necessary is according to a first embodiment of the bipolar battery concerning this invention, to laminate the n electrode layered products 9 and the n+1 electron conductive layer 11 by turns, and just to arrange the electron conductive layer 11 to the outermost layer.

[0027]

The number of times of lamination of an electrode layered product is adjusted according to the voltage for which it asks. Even if it makes thickness of a cell thin as much as possible, as long as sufficient output is securable, the number of times of lamination of a bipolar electrode may be lessened.

โกกวล

Next, a second embodiment of the bipolar battery concerning this invention, In a bipolar battery with the structure which laminated the electrode layered product which has a positive electrode layer on the field of one of the two of polymer electrolyte membrane of one sheet, and has a negative electrode layer on the field of another side via the layer of electron conductivity, A electron conductive layer has a layer which mainly contains an insulating polymer and a electron conductive filler at least in one side of a metal layer.

[0029]

In a second embodiment of the bipolar battery concerning this invention. Even when using a metal layer (for example, metallic foil) for a part of electron conductive layer, do the same operation effect so with a first embodiment having explained, and also for the corrosion-resistant problem evasion by the side of a cathode, etc., As a electron conductive layer, the layered product of a metal layer and the layer which the insulating polymer was made to fill up with a electron conductive filler can be used. It is effectively utilizable as one sort of a metal layer by what (it arranges) the copper foil which cannot carry out normal use also laminates by this the layer which made the insulating polymer fill up the positive electrode layer side with a electron conductive filler like carbon black for. Therefore, even if it is a case where the layered product of such a metal layer and the layer which the insulating polymer was made to fill up with a electron conductive filler is used as a electron conductive layer, the cell of high energy density and high power density can be constituted.

The layer which mainly contains the insulating polymer in a second embodiment of the bipolar battery concerning this invention provided at least in one side of a metal layer, and a electron conductive filler (among this Description) only — a principal layer — saying — since it is the same composition as the electron conductive layer explained in the first form of this invention which carried out point \*\*, explanation here is omitted.

As for electron conductive layer thickness, also in a second embodiment of the bipolar battery concerning this invention, it is desirable that it is comparable as the electron conductive layer thickness kicked in the first mode of this invention from a viewpoint of enlarging the energy density of a cell and power density by thin film-ization. Therefore, although main layer thickness is based also on the thickness of a metal layer, it is preferably desirable that it is the range of 0.5-10 micrometers more preferably 0.1-20 micrometers. When main layer thickness is less than 0.1 micrometer, a merit becomes small in energy density. Main layer thickness here shall say the thickness of the sum total of these both sides, when providing the thickness in both sides of a metal layer again, in providing in one side of a metal layer.

The principal layer which is one of the constituent features of a electron conductive layer does not have strong electronic conduction resistance to a plane direction, where a electron conductive filler is distributed in an insulating polymer, and its ion conduction resistance should be dramatically strong just.
[0033]

Among these, although it can be conventionally used as the above-mentioned electron conductive filler, choosing from a publicly known thing suitably, it is carbon black preferably. From the dispersibility into the insulating polymer of the above-mentioned principal layer, this is a point which can perform various surface modifications, and can be said for carbon black to be preferred. By using carbon black for the electron conductive filler of a principal layer, the purpose (high-energy-density-izing, high-power-density-izing) can be more easily attained by low cost.

Especially as the above-mentioned insulating polymer, it is not restricted, and it can be used, choosing from what [ as shown below ] is conventionally publicly known suitably for example.

(1) A thermoplastic insulating polymer can be used. As this thermoplastic insulating polymer, polyethylene, polypropylene, polystyrene, etc. can use it conveniently, for example.
[0036]

As a principal layer and a method of manufacturing a electron conductive layer further, the principal layer which carried out kneading dispersion to the insulating polymer which carried out melting of the electron conductive filler and which was made to thin-film-ize by extrusion-molding processing can be formed using the insulating polymer of this thermoplasticity, for example. The thin film of this principal layer can form a electron conductive layer in laminating to one side or both sides of a metal layer. Or although the thin film of a principal layer can be stuck and laminated to an electrode (electrode formed on the electrolyte layer in detail) and a electron conductive layer and also an electrode layered product can also be formed in laminating a metal layer on the thin film of this principal layer after that, it should not be restricted to these methods at all. [0037]

(2) What the above-mentioned insulating polymer and a electron conductive filler can distribute to both solvents is usable. The insulating polymer of this solvent dispersibility does not affect battery capacity, and just distributes it to a solvent. As an insulating polymer of this solvent dispersibility, a neoprene/toluene system, and various fluoroelastomers / methyl-ethyl-ketone systems can be used, for example.

[0038]

Using the insulating polymer of this solvent dispersibility, as a principal layer and a method of manufacturing a electron conductive layer further, For example, the stock slurry for principal layers which distributes this insulating polymer and a electron conductive filler to a solvent is prepared, The thin film of a principal layer can be produced by applying with direct spray coating and a doctor blade on the metal layer laminated on the electrode (electrode formed on the electrolyte layer in detail) which formed this stock slurry for principal layers in both sides of the electrolyte layer which has fixed intensity, for example, respectively, or this electrode etc. What is necessary is to dry and just to produce the thin film of a principal layer, after applying the stock slurry for principal layers and laminating a metal layer when the thin film of a principal layer is produced by applying with direct spray coating and a doctor blade on an electrode etc. Thereby, a electron conductive layer and also an electrode layered product can be formed collectively.

A electron conductive layer can be formed by applying this stock slurry for principal layers to one side or both sides of a metal layer, drying a solvent, and producing the thin film of a principal layer. In this case, the electrode layered product

mentioned later can also be formed in sticking and laminating the field by the side of the principal layer (or metal layer) of this electron conductive layer to an electrode (electrode formed on the electrolyte layer in detail).

This stock slurry for principal layers is applied to a glass plate or a PET film, After drying a solvent and producing a thin film, this thin film is stuck on an electrode (electrode formed on the electrolyte layer in detail), and while forming the thin film of a principal layer, a electron conductive layer (further electrode layered product) can also be formed by removing a glass plate and a PET film after that.

[0041]

the content of electron conductive fillers, such as carbon black in the principal layer obtained also in which process method of of the above (1) and (2) here, — desirable — 5 – 50 mass % — so that it may become the range of 10 – 30 mass % more preferably, It is desirable to adjust the loadings of the electron conductive filler in a raw material. When the content of a electron conductive filler is less than 5 mass %, resistance becomes large, and in exceeding 50 mass %, it becomes difficult to build a uniform thin film.

[0042]

The size of a electron conductive filler may be the same as that of the electron conductive filler in a 1st embodiment described previously.

[0043]

As a metal layer (material) which is one of the constituent features of the above-mentioned electron conductive layer, Not the thing restricted especially but what is conventionally publicly known can be used, for example, aluminium foil, stainless steel foil, the clad plate of nickel and aluminum, the clad plate of copper and aluminum, or the plating material of the combination of these metal can be used preferably. A surface of metal may be made to cover aluminum. What pasted two or more metallic foils together depending on the case may be used. It is preferred to use aluminium foil as a metal layer from viewpoints of corrosion resistance, the ease of making, economical efficiency, etc.

[0044]

The thickness of a metal layer also makes thin charge collector slack electron conductivity layer thickness in a bipolar battery, and can say it that it is desirable that it is the range of 3–10 micrometers more preferably 1–20 micrometers from a viewpoint which enlarges the energy density of a cell, and power density. When the thickness of a metal layer is less than 1 micrometer, it is a manufacture top problem, and in exceeding 20 micrometers, a merit becomes small like an energy density of a cell.

[0045]

Although the ratio in particular of the thickness of the above-mentioned principal layer and a metal layer is not restricted, it should just be about 10:1-1:20.

[0046]

Hereafter, the typical working example of the second form of this invention is briefly described using Drawings. However, it cannot be overemphasized that this invention is not what should be restricted to these embodiments in any way.

Drawing 2 shows the partial section schematic view which expressed typically second one typical embodiment of the bipolar battery concerning this invention. As shown in <u>drawing 2</u>, the second bipolar battery 21 of this invention, It has the positive electrode layer 25 on the field of one of the two of polymer electrolyte membrane (high polymer electrolyte layer 23) of one sheet, It is what has the structure which laminated the electrode layered product 29 which has the negative electrode layer 27 via the electron conductive layer 31 on the field of another side, It has the layer (layer which has electron conductive) 31b in which the electron conductive layer 31 mainly contains an insulating polymer and a electron conductive filler in at least one side of the metal layer 31a, and the field where the inside of a figure touches the positive electrode layer 25. In other words, it is a field of 1 of the electron conductive layer 31 as a charge collector (in <u>drawing 2</u>.). The positive electrode layer 25 is arranged to the layer 31b side which has electron conductivity, and it can be said that it has the structure which carried out the plural laminates of bipolar electrode 29' of the structure which has arranged the negative electrode layer 27 via polymer electrody layer 25, the electron conductive layer (charge collector) 31 (the layer 31b, the metal layer 31a which have electron conductivity), and the negative electrode layer 27 have the structure laminated in this order.

[0048]

What is necessary is according to a second embodiment of the bipolar battery concerning this invention, to laminate the n electrode layered products 29 and the n+1 electron conductive layer 31 by turns, and just to arrange the electron conductive layer 31 to the outermost layer.

[0049]

The number of times of lamination of an electrode layered product is adjusted according to the voltage for which it asks. Even if it makes thickness of a cell thin as much as possible, as long as sufficient output is securable, the number of times of lamination of a bipolar electrode may be lessened.

In order to prevent the shock from the outside, and environmental degradation in the bipolar battery concerning this invention, it is good to accommodate the main part of a bipolar battery (refer to drawing 1 and 2.) laminated by the sheet shaped in battery armor material or a cell case (not shown). The inside of the battery armor material which closes the main part of a bipolar battery, or a cell case, What covered inner surfaces (both sides), such as aluminum, stainless steel, nickel, and copper, with conventionally publicly known battery armor material like the metal-polymer compound laminate film laminated with insulators, such as a polypropylene film, is preferred.

[0051]

The bipolar battery concerning this invention is used for the rechargeable lithium-ion battery with which charge and discharge are carried by movement of a lithium ion. however, if the effect of Hitoshi Kougami of a battery characteristic is acquired, it will not bar applying to the cell of other kinds.

[0052]

Next, the component of the bipolar battery of this invention is explained individually.

[0053]

[A electron conductive layer]

A electron conductive layer is constituent features by which it is characterized [ of this invention ], and is as having explained in the first and a second embodiment which carried out point \*\*.

Namely, the electron conductive layer which can be used for this invention, A electron conductive layer is characterized by mainly including an insulating polymer and a electron conductive filler, and where a electron conductive filler is distributed in an insulating polymer, ion conduction resistance should just set [ very ] and kick greatly [ electronic conduction resistance ] to a plane direction.

[0055]

As a electron conductive filler, although carbon black etc. can be used suitably, it is not restricted to this. The quantity of this electron conductive filler, especially carbon black has good 5 - 50 mass %, and is ten to 30 mass % more preferably. [0.056]

\*\*1. Thermoplastic polymers, such as polyethylene, polypropylene, and polystyrene, can be used for an insulating polymer. [0057]

\*\*2. What an insulating polymer and a electron conductive filler can distribute to a solvent can be used. Here, as an insulating polymer which can be distributed to a solvent, what is necessary is to attain to battery capacity in influence, and for there to be nothing and for \*\* just to be able to distribute to a solvent, and a neoprene/toluene system, and various fluoroelastomers / methyl-ethyl-ketone systems can be used.

[0058]

[Positive electrode layer (it is also called a positive active material layer)]

A positive electrode layer contains positive active material. In addition, in order to improve the polymer, lithium salt, and electron conductivity which have polarity as a polymer electrolyte, a conductive auxiliary, a binder, etc. may be contained. [0059]

When using a solid polymer electrolyte for a high polymer electrolyte layer, it is desirable to contain the solid polymer electrolyte also in a positive electrode layer. By filling up the void between the positive active material in a positive electrode layer with a solid polymer electrolyte, it is because the ion conduction in a positive electrode layer becomes smooth and output improvement as the whole bipolar battery can be planned.

On the other hand, when using a polymer gel electrolyte for a high polymer electrolyte layer, the polymer electrolyte does not need to be contained in the positive electrode layer, and the conventionally publicly known binder which ties up positive—active—material particles should just be contained.

[0061]

As positive active material, the multiple oxide of the transition metal and lithium in which the lithium ion battery of a solution system is also used can be used. Specifically, Li-Fe system multiple oxides, such as Li-Mn system multiple oxides, such as Li-nickel system multiple oxides, such as Li-Oo system multiple oxides, such as LiCoO<sub>2</sub>, and LiNiO<sub>2</sub>, and spinel LiMn<sub>2</sub>O<sub>4</sub>, and LiFeO<sub>2</sub>, etc. are mentioned. In addition, the phosphoric acid compound and sulfated compound;V<sub>2</sub>O<sub>5</sub> of transition metals, such as LiFePO<sub>4</sub>, and lithium, Transition metal oxides and sulfides, such as MnO<sub>2</sub>, TiS<sub>2</sub>, MoS<sub>2</sub>, and MoO<sub>3</sub>;P bO<sub>2</sub>, AgO, NiOOH, etc. are mentioned.

[0062]

The particle diameter of positive active material is good to use a thing smaller than the particle diameter for which an electrolyte is used with the lithium ion battery of the solution type which is not a solid and which is generally used, in order to reduce the electrode resistance of a bipolar battery. Specifically, it is good in the mean particle diameter of positive active material being 0.1-5 micrometers.

[0063]

Especially as a polymer for solid polymer electrolytes, it is not limited and polyethylene oxide (PEO), polypropylene oxide (PPO), these copolymers, etc. are mentioned. \*\* -- a polyalkylene oxide system polymer [ like ] may dissolve well lithium salt, such as LiBF<sub>4</sub>, LiPF<sub>6</sub>. LiN(SO<sub>2</sub>CF<sub>3</sub>) 2, and LiN(SO<sub>2</sub>C<sub>2</sub>F<sub>5</sub>) 2. The outstanding mechanical strength is revealed by forming the structure of cross linkage. A solid polymer electrolyte is contained in either [ at least ] a positive active material layer or a negative electrode active material layer in this invention. However, in order to raise the battery characteristic of a bipolar battery more, being contained to both sides is preferred.

[0064]

As lithium salt, LiBF<sub>4</sub>, LiPF<sub>6</sub>, LiN(SO<sub>2</sub>CF<sub>3</sub>)  $_2$ , LiN(SO<sub>2</sub>C<sub>2</sub>F<sub>5</sub>)  $_2$ , or these mixtures can be used. However, it is not necessarily restricted to these.

[0065]

Acetylene black, carbon black, graphite, etc. are mentioned as a conductive auxiliary. However, it is not necessarily restricted to these.

**F0066** 

Polyvinylidene fluoride (PVDF), SBR, polyimide, etc. can be used as the above-mentioned binder. However, it is not necessarily restricted to these.

[0067]

The loadings of the positive active material in a positive electrode layer, a solid polymer electrolyte, lithium salt, a conductive auxiliary, a binder, etc. should be determined in consideration of the purposes of using a cell (output serious consideration, energy serious consideration, etc.), and ion conductivity.

[0068]

For example, if the case where the loadings are determined is taken for an example when using a solid polymer electrolyte and there are too few loadings of the solid polymer electrolyte in a positive electrode layer, ion conduction resistance and ionic diffusion resistance within a positive electrode layer will become large, and battery capacity will fall. On the other hand, if there are too many loadings of the solid polymer electrolyte in a positive electrode layer, the energy density of a cell will fall. Therefore, what is necessary is just to determine the amount of solid polymer electrolytes corresponding to the purpose in consideration of these factors. What is necessary is just to determine suitably similarly about the loadings of other constituents.

[0069]

The case where the bipolar battery which gives priority to cell reaction nature here using the solid polymer electrolyte (ionic conductivity:  $10^{-5} - 10^{-4}$ S/cm) of an actual condition level is manufactured is considered concretely. \*\* — in order to obtain the bipolar battery which has the feature [ like ], a conductive auxiliary is made more, or bulk density of an active material is lowered, and the electronic conduction resistance between active material particles is maintained lowness. A cavity part is increased simultaneously and this cavity part is filled up with a solid polymer electrolyte. \*\* — it is good for processing [ like ] to raise the rate of a solid polymer electrolyte. [0070]

The thickness in particular of a positive electrode layer should not be limited, and as loadings were described, it should be determined in consideration of the purposes of using a cell (output serious consideration, energy serious consideration, etc.), and ion conductivity. The thickness of a general positive active material layer is about 10-500 micrometers.

[Negative electrode layer]

A negative electrode layer contains negative electrode active material. In addition, in order to improve a polymer electrolyte and ion conductivity and to improve lithium salt and electron conductivity, a conductive auxiliary, a binder, etc. may be contained.

[0072]

When using a solid polymer electrolyte for a high polymer electrolyte layer, it is desirable to contain the solid polymer electrolyte also in a negative electrode layer. By filling up the void between the negative electrode active material in a negative electrode layer with a solid polymer electrolyte, it is because the ion conduction in a negative electrode layer becomes smooth and output improvement as the whole bipolar battery can be planned.

[0073] On the other hand, when using a polymer gel electrolyte for a high polymer electrolyte layer, the polymer electrolyte does not need to be contained in the negative electrode layer, and the conventionally publicly known binder which ties up negative—electrode-active-material particles should just be contained. Except the kind of negative electrode active material, since it is the same as that of the contents fundamentally indicated by the clause of the "positive electrode layer", explanation is omitted here.

[0074]

[0075]

As negative electrode active material, the negative electrode active material in which the lithium ion battery of a solution system is also used can be used. Although carbon, a metallic oxide, a lithium metal multiple oxide, etc. can be used, specifically, they are carbon or a lithium transition metal multiple oxide preferably. It is because the cell excellent in capacity and output characteristics (for example, cell voltage is made highly) can be constituted from using these. As a lithium transition metal multiple oxide, a lithium titanium multiple oxide etc. can be used, for example. As carbon, black lead, hard carbon, soft carbon, etc. can be used, for example.

[High polymer electrolyte layer]

As the above-mentioned high polymer electrolyte layer, it should not be restricted to a solid polymer electrolyte, and even if it is a polymer gel electrolyte, if liquid junction can be prevented by forming an insulating layer in the circumference of a cell layer, it can use, and also these can also be used together. An electrolyte layer can also be made into multilayer structure and the layer which changed an electrolytic kind and ingredient compounding ratio by the cathode and anode side can also be formed. When using a polymer gel electrolyte, although the ratio (mass ratio) in particular of polymer and the electrolysis solution which constitute this polymer gel electrolyte should not be restricted, its use in the range of 20:80-95:5 is common. [0076]

The above-mentioned solid polymer electrolyte layers are a polymer which has ion conductivity, and a layer which comprises lithium salt, and material will not be limited if ion conductivity is shown. As a polymer which has ion conductivity, polyethylene oxide (PEO), polypropylene oxide (PPO), and a publicly known polymer like these copolymers are mentioned. As lithium salt, LiBF<sub>4</sub>, LiPF<sub>6</sub>, LiN(SO<sub>2</sub>CF<sub>3</sub>)  $_2$ , LiN(SO<sub>2</sub>CF<sub>5</sub>)  $_2$ , or these mixtures can be used. However, it is not necessarily restricted to these. A polyalkylene oxide system polymer like PEOPPO may dissolve well lithium salt, such as LiBF<sub>4</sub>, LiPF<sub>6</sub>, LiN(SO<sub>2</sub>CF<sub>3</sub>)  $_2$ .

and  $LiN(SO_2C_2F_5)_2$ . The outstanding mechanical strength is revealed by forming the structure of cross linkage.

[0077]

A polymer gel electrolyte contains the electrolysis solution usually used for a polymer matrix with a lithium ion battery, as specified above.

[0078]

These solid polymer electrolytes or a polymer gel electrolyte may be contained also in a positive electrode layer and/or a negative electrode layer as everything but the high polymer electrolyte layer which constitutes a cell was described above, but. The polymer electrolyte which changes with the high polymer electrolyte layers, positive electrode layers, and negative electrode layers which constitute a cell may be used, the same polymer electrolyte may be used and the polymer electrolyte which changes with layers may be used.

[0079]

The thickness in particular of the high polymer electrolyte layer which constitutes a cell is not limited. However, in order to obtain a compact bipolar battery, it is preferred to make it thin as much as possible in the range which can secure the function as an electrolyte. The thickness of a general high polymer electrolyte layer is about 10-100 micrometers. [0080]

[A cathode and a negative terminal block]

What is necessary is just to use a cathode and a negative terminal block if needed. When using, it has a function as a terminal, and also the thinner one as much as possible is good from a viewpoint of slimming down, but as for each of electrodes which it comes to laminate, electrolytes, and charge collectors, since the mechanical strength is weak, it is desirable to give only the intensity which carries out \*\*\*\*\*\* support of these from both sides. The thickness of a cathode and a negative terminal block can be said for about 0.1-2 mm to be usually desirable from a viewpoint of stopping the internal resistance in a terminal area.

[0081]

The construction material usually used with a lithium ion battery can be used for the construction material of a cathode and a negative terminal block. For example, aluminum, copper, titanium, nickel, stainless steel (SUS), these alloys, etc. can be used. It is preferred to use aluminum from viewpoints of corrosion resistance, the ease of making, economical efficiency, etc. [0082]

The same construction material may be used for the construction material of a positive pole terminal board and a negative terminal block, and the thing of different construction material may be used for it. These cathodes and a negative terminal block may laminate to a multilayer that from which construction material differs.

[0083]

[A cathode and a negative electrode lead]

About a cathode and a negative electrode lead, the publicly known lead usually used with a lithium ion battery can be used. Since the portion taken out from battery armor material (cell case) does not have distance with the heat source of a car, it is preferred to cover with the heat-shrinkable tubing of heat-resistant insulation, etc. so that these may be contacted, it may be short-circuited and autoparts (especially electronic device) may not be affected.

[0084]

[Battery armor material (cell case)]

A bipolar battery is good to accommodate the whole cell layered product which is a main part of a bipolar battery in battery armor material thru/or a cell case, in order to prevent the shock from the outside at the time of using it in order to prevent the shock from the outside, and environmental degradation, and environmental degradation. The polymer-metal compound laminate film etc. which covered metal (an alloy is included), such as aluminum, stainless steel, nickel, and copper, with insulators, such as a polypropylene film, from a viewpoint of a weight saving, It is preferred by joining a part or all of the periphery in thermal melting arrival using publicly known battery armor material conventionally to have composition which stored and sealed the cell layered product. In this case, the above-mentioned cathode and a negative electrode lead should just be taken as the structure which is inserted into the above-mentioned heat sealed part, and is exposed to the exterior of the above-mentioned battery armor material. It is preferred to use the polymer-metal compound laminate film excellent in thermal conductivity etc. at the point that heat can be efficiently told from the heat source of a car, and the inside of a cell can be quickly heated to battery operation temperature.

[0085]

Next, it can be considered as the cell group which connected and constituted the above-mentioned bipolar battery from this invention. [ two or more ] That is, it becomes possible to satisfy comparatively inexpensive the demand to the cell capacity and the output for every purpose of use by considering it as the cell group which connected in series and/or in parallel and constituted the bipolar battery of this invention using at least two or more pieces.

[0086]

Concrete for example, the above-mentioned bipolar battery is connected to N piece parallel, and the bipolar battery made N piece parallel is made into M more piece series, and it stores to the battery case made of metal thru/or resin, and is considered as a cell group. Under the present circumstances, the series / the number of multiple connection of a bipolar battery are determined according to the purpose of use. For example, what is necessary is just to combine so that it can apply to the power supply for a drive of the vehicles with which high energy density and high power density are called for as large capacity power supplies, such as an electromobile (EV) and a hybrid electric vehicle (HEV). What is necessary is just to electrically connect the positive pole terminal for cell groups and a negative pole terminal, and the electrode lead of each bipolar battery using a lead etc. What is necessary is just to electrically connect using a suitable connecting member like a spacer or a bus bar, when connecting bipolar batteries in series/in parallel. However, the cell group of this invention should

not be restricted to what was explained here, and a publicly known thing can be conventionally used for it suitably. In order to form various measuring machine machines and control machinery, for example, to supervise cell voltage according to usage, it is not restricted [ provide / the connector for voltage Measurement Division, etc. ] in particular to this cell group. [0087]

In this invention, it can be considered as the vehicles which carry an above-mentioned bipolar battery and/or cell group as a power supply for a drive. The bipolar battery and/or cell group of this invention have the various characteristics as mentioned above, and are an especially compact cell. For this reason, it is suitable as power supplies for a drive, such as the vehicles with which an especially severe demand is made about an energy density and power density, for example, an electromobile, and a hybrid electric vehicle. For example, since carrying a cell group as a power supply for a drive under the seat of the body center section of an electromobile thru/or the hybrid electric vehicle can take in-company space and a large trunk room, it is convenient. However, in this invention, it may carry in the lower part of the rear trunk room instead of what should be restricted to these in any way, or if an engine is not carried like an electromobile, it can also carry in the portion etc. in which the engine ahead of the body was carried. It may be made to carry a bipolar battery not only depending on a cell group but depending on usage, and may be made to carry in this invention combining these cell groups and a bipolar battery. Although an above-mentioned electromobile and hybrid electric vehicle are preferred as vehicles which can carry the bipolar battery and/or cell group of this invention as a power supply for a drive, it is not restricted to these.

Especially as a manufacturing method of the bipolar battery of this invention, it should not be restricted and various kinds of publicly known methods can be used suitably conventionally. Below, it explains briefly. f00891

(1) Formation of a electron conductive layer

The electron conductive layer which mainly contains an insulating polymer and a electron conductive filler is obtained by the following.

[0090]

1When using thermoplastic polymers, such as polyethylene, polypropylene, and polystyrene, for an insulating polymer, kneading dispersion is carried out to the insulating polymer which carried out melting of the electron conductive filler, and it is obtained by thin-film-izing by extrusion-molding processing.

[0091]

2When an insulating polymer and a electron conductive filler use what can be distributed to a solvent, it is obtained by producing to an electrode the slurry which dissolves an insulating polymer and a electron conductive filler in a solvent by spreading etc. with direct spray coating and a doctor blade. Or the above-mentioned slurry is applied on support materials, such as a glass plate and a PET film, and it is obtained by drying a solvent and producing a thin film. \*\*2. The \*\* case forms beforehand the electrode formed henceforth [ (2) ] on suitable support materials (a glass plate, a PET film, etc.) thru/or an electrolyte layer, and is a electron conductive layer on this electrode 2 What is necessary is just to form by \*\*\*\*\*\*. That is, the procedure of explaining below is the above 1. It is a procedure of a \*\* case and is 2. It carries out to a \*\* case by replacing these formation order, spreading and lamination subjects thru/or spreading and a subject-ed [ lamination ]. However, it will be 1 if it removes forming in an electrode the electron conductive layer which changes having formed the electrode in the existing charge collector to a charge collector when a cell is constituted, as shown in already explained drawing 1 and 2. A cell can be formed similarly. Therefore, 2 Since it is carrying out in the working example about the manufacture procedure of the cell using \*\*\*\*\*\*\*\*\*\*, explanation here is omitted.

(2) Spreading of the constituent for cathodes

The constituent for cathodes is obtained as a slurry (slurry for cathodes), and is usually applied on FAIRUMU, such as PET. In addition, the above-mentioned \*\*1. The electron conductive layer come out of and produced may be prepared, and it may apply to the field of one of these.

[0093]

Including positive active material, as other ingredients, the slurry for cathodes has a raw material of a conductive auxiliary, a binder, a polymerization initiator, and the polymer for polymer electrolytes, lithium salt, an arbitrary solvent, etc., and is contained. In using a polymer gel electrolyte for a high polymer electrolyte layer, The conductive auxiliary for improving the conventionally publicly known binder which ties up positive-active-material particles, and electron conductivity, the solvent, etc. should just be contained, and host polymer, an electrolysis solution, lithium salt of the raw material of the polymer gel electrolyte, etc. do not need to be contained.

[0094]

As for the polymer raw material of a polymer electrolyte, it is preferred for PEO, PPO(s), these copolymers, etc. to be mentioned and to have the functional groups (carbon-carbon double bond etc.) of cross-linking in intramolecular. A mechanical strength improves by constructing a bridge in a polymer gel electrolyte using the functional group of this cross-linking.

[0095]

The compound mentioned above can be used about positive active material, a conductive auxiliary, a binder, and lithium salt. [0096]

It is necessary to choose a polymerization initiator according to the compound to polymerize. For example, azobisisobutyronitrile is mentioned as benzyl dimethyl ketal and a thermal polymerization initiator as a photopolymerization initiator.

[0097]

Solvents, such as NMP, are chosen according to the kind of slurry for cathodes. [0098]

What is necessary is just to adjust the addition of the polymer raw material of positive active material, lithium salt, a conductive auxiliary, a binder, and a polymer electrolyte, etc. according to the purpose of a bipolar battery, etc., and it should just add the quantity usually used. As for the addition of a polymerization initiator, a \*\*\*\*\*\*\* decision of the polymer raw material of a polymer electrolyte is made. Usually, it is a 0.01 - 1 mass % grade to a polymer raw material. [0099]

(3) Formation of a positive electrode layer

The electron conductive layer to which the slurry for cathodes was applied is dried, and the solvent contained is removed. Crosslinking reaction is advanced depending on the slurry for cathodes, and, simultaneously with it, the mechanical strength of a solid polymer electrolyte may be raised. The desiccation can use a vacuum dryer etc. Although the conditions of desiccation are determined according to the applied slurry for cathodes and it cannot specify uniquely, it is usually 1 to 6 hours at 80-120 \*\*.

[0100]

(4) Spreading of the constituent for anodes

The constituent for anodes is obtained as a slurry (slurry for cathodes), and is usually applied on FAIRUMU, such as PET. In addition, the constituent for anodes (slurry for anodes) containing negative electrode active material may be applied to the field where the positive electrode layer was applied, and the field of the electron conductive layer of an opposite hand. [0101]

Including negative electrode active material, as other ingredients, the slurry for anodes has a raw material of a conductive auxiliary, a binder, a polymerization initiator, and the polymer for polymer electrolytes, lithium salt, an arbitrary solvent, etc., and is contained. About the raw material and addition which are used, since it is the same as that of explanation by the clause of "spreading of the constituent for (1) cathodes", explanation is omitted here.

(5) Formation of a negative electrode layer

The electron conductive layer to which the slurry for anodes was applied is dried, and the solvent contained is removed. Crosslinking reaction is advanced depending on the slurry for anodes, and, simultaneously with it, the mechanical strength of a polymer electrolyte may be raised. A bipolar electrode is completed according to this work. The desiccation can use a vacuum dryer etc. Although the conditions of desiccation are determined according to the applied slurry for anodes and it cannot specify uniquely, it is usually 1 to 6 hours at 80-120 \*\*.

(6) Lamination with a bipolar electrode and a high polymer electrolyte layer Separately, the high polymer electrolyte layer laminated by inter-electrode is prepared. [0104]

When using a polymer solid electrolyte layer, it is manufactured by stiffening the solution which was made to dissolve the high-cost-of-raw-materials molecule of a solid polymer electrolyte, lithium salt, etc. in a solvent like NMP, and was prepared, for example. When using a polymer gel electrolyte layer, it is manufactured by, for example, polymerizing simultaneously with stoving the pregel solution which consists of host polymer, an electrolysis solution and lithium salt, a polymerization initiator, etc. under an inert atmosphere as a raw material of a polymer gel electrolyte (crosslinking reaction is promoted). [0105]

The thickness of these high polymer electrolyte layers is controllable using a spacer. When using a photopolymerization initiator, it is good to slush into the gap of a light transmittance state, to irradiate with ultraviolet rays, to advance crosslinking reaction, and to produce a film. However, of course, it is not limited to this method. According to the kind of polymerization initiator, radiation initiated polymerization, an electron beam polymerization, thermal polymerization, etc. are used properly. About the high-cost-of-raw-materials molecule of the polymer electrolyte to be used, an electrolysis solution, lithium salt, a polymerization initiator, and each loadings, since it is as having mentioned above, explanation is omitted here. [0106]

After carrying out stoving of the polymer solid electrolyte layer to the positive/negative electrode produced as mentioned above enough under a high vacuum, two or more each is started for a positive/negative electrode and a high polymer electrolyte layer in suitable size. Predetermined number \*\*\*\*\*\*\*\*\* and a layer built cell are produced for the bipolar electrode and high polymer electrolyte layer which were started. The number of laminations is determined in consideration of the battery characteristic for which a bipolar battery is asked. On the high polymer electrolyte layer of the outermost layer, an electrode is arranged, respectively. In the outermost layer by the side of a cathode, the electrode in which only the positive electrode layer was formed on the electron conductive layer is arranged. In the outermost layer by the side of an anode, the electrode in which only the negative electrode layer was formed on the electron conductive layer is arranged. As for the stage of making a positive/negative electrode and a high polymer electrolyte layer laminating, and obtaining the main part of a bipolar battery (cell layered product), it is preferred to perform from a viewpoint to prevent that moisture etc. mix in the inside of a cell under an inert atmosphere. For example, it is good to produce a bipolar battery under argon atmosphere and a nitrogen atmosphere.

[0107]

(7) Packing (completion of a cell)

Finally, a positive pole terminal board and a negative terminal block are installed, respectively on the electron conductive layer of both the outermost layers of the main part of a bipolar battery (cell layered product), and a positive electrode lead and a negative electrode lead are further joined and (it electrically connects) taken out to this positive pole terminal board

and a negative terminal block. Although it should not be restricted especially as a joining method of a positive electrode lead and a negative electrode lead, and ultrasonic welding with low welding temperature, etc. can use suitably, it should not be limited to this and a publicly known joining method can be used suitably conventionally.

[0108]

The whole cell layered product is closed with battery armor material thru/or a cell case, in order to prevent the shock from the outside, and environmental degradation, and a bipolar battery is completed. The metal (aluminum, stainless steel, nickel, copper, etc.) with which the inner surface was covered by insulators, such as a polypropylene film, is preferred for the construction material of battery armor material (cell case).

[0109]

[Working example]

The effect of this invention is explained using the following working examples and comparative examples. However, technical scope of this invention is not limited to the following working examples.

[០110]

Hereafter, the working example and comparative example of this invention are explained.

[0111]

As a polymer electrolyte, the raw material of the network polymer of a polyether form compounded in accordance with the method of document is used (J. Electrochem. Soc., 145 (1998) 1521.), As lithium salt, LiN(SO<sub>2</sub>C<sub>2</sub>F<sub>5</sub>) <sub>2</sub> (it carries out abbreviated [ of this ] to BETI henceforth) was used.

[0112]

(1) Production of polymer electrolyte membrane

First, production of polymer electrolyte membrane was performed as follows. After preparing benzyl dimethyl ketal as 26 mass % and a photopolymerization initiator and preparing the \*\*\*\* solution for dry acetonitrile for BETI as 0.1 mass [ of a polymer raw material ] %, in addition a solvent as 53 mass % and lithium salt, acetonitrile was removed for the above-mentioned polymer raw material by vacuum distillation. Thickness was specified using the Teflon (registered trademark) spacer, the high solution of this viscosity was filled between glass substrates, it irradiated with ultraviolet rays for 20 minutes, and photopolymerization (bridge construction) was carried out. The film was taken out, it put into the vacuum housing, stoving was carried out under a 12-hour high vacuum at 90 \*\*, and residual water and a solvent were removed. The thickness of the produced polymer electrolyte membrane was 20 micrometers.

[0113]

(2) Production of a negative electrode layer

Production of the negative electrode layer was performed as follows. Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub> was used as negative electrode active material. This electrode active material 28 mass %, acetylene black 3 mass %, and the above-mentioned polymer raw material 17 mass %, 0.1 mass % A polymer raw material adds azobisisobutyronitrile for BETI as 8 mass % and a thermal polymerization initiator, NMP was 44 mass % Added to this as a solvent, it fully agitated, the slurry was prepared, it applied by the coating machine on the PET philharmonic, stoving was carried out at 90 \*\* with the vacuum dryer for 2 hours or more, and the anode was produced. The thickness of the produced negative electrode active material layer was 22 micrometers.

[0114]

(3) Production of a positive electrode layer

Production of the positive electrode layer was performed as follows. The LiMn<sub>2</sub>O<sub>4</sub> positive electrode layer was used as positive active material. In detail LiMn<sub>2</sub>O<sub>4</sub> with a mean particle diameter of 2 micrometers of 29 mass %, Acetylene black of 8.7 mass %, the above-mentioned polymer raw material of 17 mass %, 0.1 mass % A polymer raw material adds azobisisobutyronitrile for BETI as 7.3 mass % and a thermal polymerization initiator, NMP was 41 mass % Added to this as a solvent, it fully agitated, the slurry was prepared, it applied by the coating machine on the PET philharmonic, stoving was carried out at 90 \*\* with the vacuum dryer for 2 hours or more, and the cathode was produced. The thickness of the produced positive active material layer was 20 micrometers.

[0115]

In the following working examples and comparative examples, production and an experiment were altogether conducted on condition of the following.

[0116]

\*\*1. Size of the polar zone: 30 mm x 30 mm.

\*\*2. The number of electrode lamination cells (cell layer): Ten,

\*\*3. Charge-and-discharge conditions: Discharge considered 0.2C constant current and 27V constant potential charge as 10V cutoff in 0.1C for a total of 10 hours.

[0117]

Working example 1

\*\*1. adding and carrying out melt kneading of the carbon black (it is about 30 mass % to the total amount of polyethylene and carbon black) to polyethylene, using a heat press roll, and lengthening -- 10 micrometers in thickness -- it was used, having carried out electron conductive film formation.

[0118]

With the described method, the electrolyte membrane, the positive electrode layer, and the negative electrode layer were produced, respectively. On copper foil, a negative electrode layer / high polymer electrolyte layer / positive electrode layer is formed, and it is 1 on a positive electrode layer. \*\*\*\*\*\*\*\*\*\*\*\*\*\*\* has been arranged and the negative electrode layer / high polymer electrolyte layer / positive electrode layer has been further arranged on it. This was repeated 10 times and aluminum

foil has been arranged on the last positive electrode layer.

[0119]

The tab (terminal lead) of nickel was attached to copper foil and aluminum foil, vacuum enclosure was carried out and the polar zone was made the pack of the laminate film of aluminum with the cell.

[0120]

As this example showed, in this invention, it may replace with this electron conductive layer, and the conventional charge collector may be adopted as the electron conductive layer of the electrode of the outermost layer of a cell. In a first embodiment that does not use a metal layer in particular, it is because it is useful at the point that a terminal lead is easily connectable.

[0121]

Working example 2

1Distribute a neoprene and carbon black in toluene (making 40 mass % into carbon black in solid content about 20 mass %), and on a PET film, carry out spray coating and hot air drying is carried out, The 5-micrometer-thick electron conductive film was produced, it used instead of the electron conductive film of the working example 1, and the same bipolar battery as the working example 1 was constituted.

[0122]

Working example 3

The electron conductive film of the working example 2 was formed on 4-micrometer-thick copper foil, the field by the side of the anode of the working example 1 was made into copper foil, this electronic transmission nature film has been arranged, and the same bipolar battery as the working example 1 was constituted.

[0123]

Comparative example 1

Instead of the electron conductive film of the working example 1, as shown in <u>drawing 3</u>, 20-micrometer-thick aluminum foil (the first current collecting foil 51a) was used for the positive electrode layer side, 15-micrometer-thick copper foil (the second current collecting foil 51b) was used and laminated to the negative electrode layer side, and the same bipolar battery as the working example 1 was constituted.

[0124]

The energy density of each cell was calculated by making the produced bipolar battery into charge-and-discharge conditions by discharge having carried out the charge and discharge of 0.2C constant current and the 27V constant potential charge by 10V cutoff in 0.1C for a total of 10 hours, the ratio was taken to the value of the cell of the comparative example 1, and the result was shown in Table 1. By using the layer which carried out distributed restoration of the electron conductive filler into the insulating polymer in the electron conductive layer which separates between the cells of a bipolar battery (between cell layers) so that the result of Table 1 may show, The energy density of a bipolar battery can be raised compared with the case where a metallic foil is used as a charge collector.

[0125]

[Table 1]

バイポーラ電池	エネルギー密度比
実施例 1	1. 30
実施例2	1.39
実施例3	1. 32
比較例1	1. 00

[Brief Description of the Drawings]

[Drawing 1] The partial section schematic view which expressed typically a first embodiment of the bipolar battery concerning this invention is shown.

Drawing 2] The partial section schematic view which expressed typically second one typical embodiment of the bipolar battery concerning this invention is shown.

[Drawing 3]It is a bipolar battery using the conventional charge collector, and the partial section schematic view which expressed typically the bipolar battery produced by the comparative example 1 is shown.

[Explanations of letters or numerals]

- 1. 21. a 41 -- bipolar battery.
- 3, 23, 43 -- polymer electrolyte membrane (high polymer electrolyte layer),
- 5, 25, a 45 -- positive electrode layer,
- 7, 27, a 47 -- negative electrode layer,
- 9, 29, a 49 -- cell layer,

9, 29, 49 -- Bipolar electrode

11, a 31 -- electron conductivity layer,

31a -- Metal layer,

31b -- Layer which has electron conductivity,

51 -- current collecting foil,

51a -- The first current collecting foil,

51b -- The second current collecting foil.

[Translation done.]